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The Behavior of Actinides in the Environments

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Summary

A review of actinide behavior in the environment is presented with emphasis on chemical, physical, and biological factors that influence actinide mobility in ecosystems Available data from terrestrial and fresh water ecosystems suggest that physical processes which result in the transport of soils and sediments dominate in the translational movement of plutonium and, as well, dominate in the transport of this element through lower trophic levels. Exceptions to that statement occur in arctic ecosystems and in deep oceans Regardless of mode of transport, plutonium levels in higher trophic levels including man are very low indicating the low solubility of this element in the environment. Very few data on the behavior of the other actinides in the environment are currently available although theoretical considerations and limited laboratory experiments suggest that many of the actinides are more mobile than plutonium

Introduction

In the actinide series, the elements of greatest interest as environmental contaminants are uranium, neptunium, plutonium, americium, and curium because the presence of these elements at relatively high concentrations in ecosystems would represent potential health problems The purpose of this paper is to review current knowledge on the distribution and transport of those elements in terrestrial and aquatic environments under a variety of site and source conditions Based on available data, none of the actinides discussed herein are currently present, in an available form in the general environment, at concentrations considered to be a health hazard Among those elements, only uranium occurs naturally in any significant quantity The natural isotopes of uranium are so long-lived that their specific activity is very low, thus chemical toxicity involving kidney damage takes precedent over radiotoxicity in considerations of health [1] However, the isotopes ^{232}U and ^{233}U, which have higher specific activities, can be produced in the thorium breeder cycle and could represent a potential environmental health problem [2]

With current reactor technology, the isotopes of neptunium, plutonium, americium, and curium are formed during neutron bombardment of uranium through a complicated series of neutron capture and radioactive decay reactions (Fig 1, [3]) The importance of those elements as environmental contaminants depends upon factors related to their production, half life, chemistry, mode of dispersion, and biological availability The transuranic nuclides which may be of interest as environmental contaminants are listed in Table 1

Neptunium 237, by nature of its long half life, is the only isotope of neptunium that could be considered a possible long term hazard in the environment Until recently, very little consideration was given to ^{237}Np as a radiological problem because of its low production and relatively low

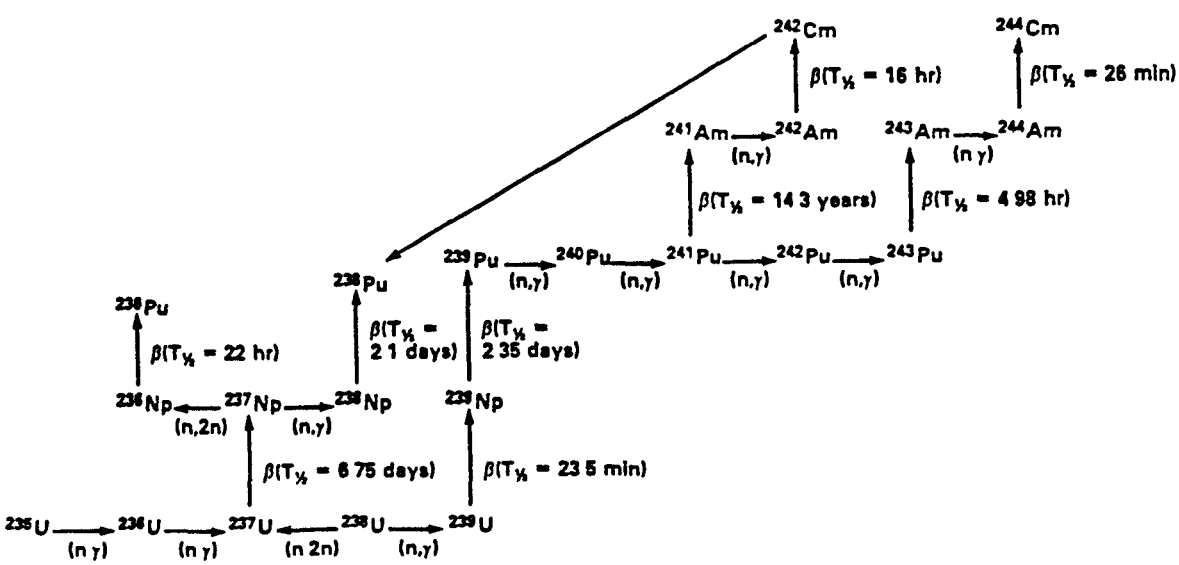

Fig 1 Formation schemes for important actinides (ref [3])

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Table 1 *Nuclear properties of long lived transuranic elements*

Element	Isotope	Emission	Half life
Neptunium	^{237}Np	α	2 1 × 10^6 y
Plutonium	^{238}Pu	α	86 4 y
	^{239}Pu	α	24,400 y
	^{240}Pu	α	6580 y
	^{241}Pu	β	13 2 y
Americium	^{241}Am	α	458 y
Curium	^{244}Cm	α	17 6 y

specific activity However, recent data indicate that neptunium is the most mobile of the subject group of transuranic elements in ecosystems and mammalian organisms [4, 5] This reported mobility could be a compensating factor for the low production and low specific activity of ^{237}Np when considering the potential radiological consequences of release to the environment

Plutonium has been produced in greater quantity than any other transuranic element Plutonium has also been the subject of the most biological and ecological research because atmospheric weapons testing, routine waste disposal and various accidents have made it more prevalent than the other subject elements in the environment The four isotopes of plutonium (Table 1) that are of most concern as environmental contaminants are ^{238}Pu, ^{239}Pu, ^{240}Pu and ^{241}Pu Plutonium chemistry is complicated because this element can form relatively stable compounds in several oxidation states under various environmental and biological conditions

Americium 241, with a half-life of over 450 years is the second most prevalent transuranic element in the environment and results primarily from nuclear weapons testing The complete decay of ^{241}Pu from present worldwide fallout should produce an amount of ^{241}Am about equivalent to ^{239,240}Pu Under environmental conditions, the chemistry of americium is relatively simple with only the Am(III) oxidation state being of importance in the environment [6]

Curium is the least important of the transuranic elements as a gobal environmental contaminant because very little of its longer lived isotope, ^{244}Cm, has been released to the environment through weapons testing However, this element is significant in power reactor wastes [7] With a half life of 18 years, ^{244}Cm can persist in the environment for several human generations As with americium, only one oxidation state, Cm(III), is important under environmental conditions [6]

All of the transuranic elements are considered to be boneseekers, when inhaled or ingested, with about equal distribution between the skeleton and the liver [8] In experimental animals receiving large radiological doses of transuranic elements, the principal cause of death has been from bone cancer [9, 10]

Sources and distribution in the environment

Only uranium among the actinides of interest occurs naturally in the environment in easily measureable concentrations It is essentially ubiquitous and concentrations in soil range from 1 to 10 μg/g [11] Uranium concentrations in soil tend to reflect the base materials from which they are derived although there is a tendency for the uranium content of organic rich horizons to be greater than other horizons in a soil profile [12] Concentrations in sea water are reported in the range of 0 3 to 6 μg/l [13] with an average concentration of 3 μg/l [14] Continental surface waters range from 0 1 μg/l to 500 μg/l depending on the amount and solubility of the uranium in the parent watershed or aquifer Concentrations of uranium in deep ocean sediments range from 0 4 to 3 0 μg/g and vary inversely with the calcium carbonate content of the sediment [15]

Extremely small amounts of the transuranic elements, principally ^{239}Pu, are formed naturally by neutron capture in uranium ores [16, 17, 18] The ratio of ^{239}Pu relative to uranium under those circumstances is 3×10^{-12} Even higher occurrence ratios have been found in volcanic rock [19], however, the major sources of plutonium in the environment are due to human activities

The ubiquitous distribution of man-made plutonium is due to the detonation of nuclear weapons in the atmosphere In particular, the testing of thermonuclear devices, which began in 1952, produced the greatest amount of airborne plutonium The large energy releases in these explosions injected nuclear debris into the stratosphere where it remained long enough to be distributed globally An estimated 360,000 curies (Ci) of the isotopes ^{239,240}Pu, and after complete decay an almost equal number of curies of ^{241}Am, and smaller amounts of other transuranic elements entered terrestrial and aquatic environments by this route (Table 2, [7]) Most of those radionuclide inventories have deposited in the northern hemisphere due to the tests conducted by the United States and the Soviet Union Environmental concentrations of fallout radionuclides, including plutonium, are highest at mid latitudes with attenuated concentrations occurring at higher and lower latitudes as shown in Table 3 [20]

About 16 kCi of ^{238}Pu were dispersed in the upper atmosphere of the southern hemisphere in April 1964 when a navigational satellite with its SNAP 9A generator

Table 2 *Estimated amounts of transuranium elements that have been injected into the atmosphere* [7]

Radionuclide	Total injected kCi
^{238}Pu	24[a]
^{239}Pu	154
^{240}Pu	209
^{241}Pu	9720
^{241}Am	336[b]

[a] Represents 16 kCi from SNAP 9A and 8 curies from weapons tests

[b] Americium 241 formed on total decay of ^{241}Pu

2

Table 3 *Average latitudinal distributions of cumulative* Pu 239, 240 *and* Pu 238 *fallout* [20]

		mCi per km²		
			Pu 238	
Hemisphere	Latitude Band	Pu-239, 240	Weapons	SNAP 9A
Northern	90 – 80	(0 10 ± 0 04)[a]	(0 002 ± 0 001)	(< 0 001)
	80 – 70	0 36 ± 0 05	0 009 ± 0 001	< 0 001
	70 – 60	1 6 ± 1 0	0 038 ± 0 025	0 026 ± 0 015
	60 – 50	1 3 ± 0 2	0 031 ± 0 004	0 013 ± 0 004
	50 – 40	2 2 ± 0 5	0 053 ± 0 011	0 026 ± 0 011
	40 – 30	1 8 ± 0 6	0 042 ± 0 014	0 025 ± 0 015
	30 – 20	0 96 ± 0 07	0 023 ± 0 002	0 011 ± 0 004
	20 – 10	0 24 ± 0 10	0 006 ± 0 002	0 003 ± 0 002
	10 – 0	0 13 ± 0 06	0 003 ± 0 001	< 0 001
Southern	0 – 10	0 30 ± 0 20	0 007 ± 0 005	0 010 ± 0 007
	10 – 20	0 18 ± 0 05	0 004 ± 0 001	0 036 ± 0 021
	20 – 30	0 39 ± 0 16	0 009 ± 0 004	0 070 ± 0 042
	30 – 40	0 40 ± 0 12	0 009 ± 0 003	0 061 ± 0 020
	40 – 50	0 35 ± 0 21	0 008 ± 0 005	0 069 ± 0 038
	50 – 60	(0 20 ± 0 09)	(0 005 ± 0 002)	(0 044 ± 0 023)
	60 – 70	(0 10 ± 0 04)	(0 002 ± 0 001)	(0 022 ± 0 012)
	70 – 80	(0 03 ± 0 01)	(0 001 ± 0 001)	(0 008 ± 0 005)
	80 – 90	(0 01 ± 0 004)	(< 0 001)	(0 004 ± 0 002)

[a] Results in parentheses were derived by extrapolation, error terms are standard deviations

failed to achieve orbit and vaporized upon reentry Essentially all of the ^{238}Pu released by this event has reached the surface of the earth, about 75 percent of this material deposited in the southern hemisphere (Table 3)

Localized sources of plutonium were created by accidents involving aircraft that were carrying nuclear weapons An explosion during mid-air refueling of a U S Air Force B 52 occurred on January 16, 1956, above Palomares, Spain The high explosives in two unarmed nuclear weapons detonated on impact near Palomares scattering the plutonium into the surrounding area The plutonium was reduced to acceptable concentrations in soil by decontamination procedures [21] On January 21, 1968, another B-52 carrying unarmed nuclear weapons crashed and burned on the ice near Thule, Greenland Most of the plutonium contained in the weapons was recovered, however, an estimated 25 Ci deposited in marine sediments of Bylot Sound and an additional 1 to 5 Ci were dispersed to nearby shore areas [22]

Plutonium 239, 240 and americium 241 are the primary alpha emitters resulting from nuclear explosions, however, under longer irradiation times such as incurred in nuclear reactors, neptunium and curium will also be produced in considerable quantities [23] Although neptunium and curium are present in irradiated reactor fuel, releases of transuranic elements during chemical separation of plutonium from reactor fuel primarily involve plutonium and americium Part of the releases from such facilities has been in the form of aerosols from ventilation systems where absolute retention of particles cannot be achieved or where air cleaning equipment has failed [24, 25] Those airborne releases result mainly in localized contamination in terrestrial ecosystems Transuranic elements also have been distributed in localized terrestrial systems through release of liquid wastes in such places as the White Oak Creek flood plain at Oak Ridge, Tennessee [26], and the waste disposal canyons at Los Alamos, New Mexico [27] Dispersal of transuranic elements into aquatic environments occurs during planned releases of low level liquid wastes into river [28, 29] or marine waters [30, 31] in accordance with national and international regulations and guidelines

The distribution of transuranic elements from point sources at nuclear facilities typically produces decreasing concentrations with distance from the source As a result, much of the readily detectable material is located within the controlled boundaries of nuclear facilities In those areas and in areas with limited public access, concentrations of those elements in soils and sediment may exceed fallout levels but generally are still low with respect to levels that would constitute a hazard to animals and humans [26, 29, 31, 32, 33, 34, 35] Controlled areas such as the Nevada Test Site, where plutonium was scattered by high explosives to simulate accident conditions, may be an exception to the latter statement As much as 200 Ci may still be distributed in relatively small areas after decontamination procedures removed most of the material [36]

A range of concentrations of plutonium in soils, sediments, and water are given in Table 4 An interesting observation from the table is that the concentrations of plutonium in water are quite low compared to soils and sediments In fact in several terrestrial ecosystems which have been studied, more than 99 percent of the plutonium inventory is ultimately associated with soil and less than 1% is associated with biota (Table 5) There are unique

3

Table 4 *Plutonium in soils sediments and water*

Source and locations	Concentration ($^{239,240}Pu$) Soils and sediments[a] (pCi/g dry wgt)	Reference
Nuclear weapons testing		
Global Fallout (Soil)	$5 \times 10^{-4} - 2 \times 10^{-2}$	37
Debris (NTS, Soil)	80 – 5×10^{4}	38
Bikini Atoll (Soil)	0 5 – 400	39
Lake Michigan (Sediment)	0 09 – 0 40	40
Trinity Site Fallout (Soil)	0 02 – 0 32	35
Chemical processing		
Savannah River, S C (Soil)	6×10^{-3} – 3 7	24
Hanford, Wash (Soil)	4×10^{-3} – 0 7	41
Rocky Flats, Colo (Soil)	4×10^{-3} – 70	42
Irish Sea (Sediment)	0 3 – 50	30
Los Alamos, NM (Alluvium)	1 – 290	35
	Water, (pCi/liter)	
Nuclear weapons testing		
Enewetak Atoll (groundwater)	2×10^{-4} – 0 7	43
Lake Michigan	2×10^{-4} – 3×10^{-4}	44
Atlantic Ocean	1×10^{-4}	45
Chemical processing		
Savannah River, S C (freshwater)	2×10^{-3}	46
Savannah River, S C (treated drinking water)	1×10^{-4}	46
Irish Sea	0 05 – 0 5	30
Los Alamos, NM (treated effluent, surface and ground water, Mortandad Canyon)	0 14 – 17	47

[a] pCi = 10^{-12} curie

Table 5 *Inventory ratios for plutonium in soil at terrestrial research sites*

Site	Compartment	Reference
Los Alamos (Mortandad Canyon)	0 997	49
Oak Ridge (White Oak Creek Flood Plain)	0 999	49
Rocky Flats	> 0 99	50
Nevada Test Site	0 997	51
Trinity Site	0 99	35

ecosystems in which plutonium behavior can differ from the norm For example, fallout plutonium in arctic ecosystems can be retained by dense lichen mats that effectively delay the transfer of plutonium to soil by several years due to an effective weathering half time of plutonium from lichens of about 6 years [22, 48] Although such circumstances can have short term significance in terms of entry of plutonium into food webs (i e lichen-caribou-eskimo), the final repository of fallout plutonium in those ecosystems will also be soil In aquatic ecosystems, the transuranic elements may vary in distribution between water and sediment depending upon the chemical environment, the volume of water relative to sediment surfaces, and the size and type of sediment material In relatively shallow water such as Lake Michigan [52], Buzzards Bay [53], the Irish Sea [54], and Trombay Harbor [55] greater than 95% of the Pu is associated with sediments In the open ocean, a smaller proportion of the plutonium may be in sediments because of the long settling time of particulate matter or slow diffusion of soluble forms to great depths [45] Data for americium are limited but indicate a distribution between water and sediment that is similar to plutonium [54] A similar inference can be made for curium, however, neither curium nor neptunium have been measured in aquatic environments in sufficient amounts to provide reliable inventories

Transport and fate in the environment

In general the actinide elements show a strong association with soils and sediments That degree of association influences the concentrations that are available for abiotic and biotic transport processes In most cases the mobile or soluble fraction is small relative to the amount adsorbed on solid matter, however, solubility is highly dependent

4

upon the chemistry of the element and upon the chemical environment to which the element is exposed

One of the most important aspects of chemical behavior of the actinide elements is the stability of the various oxidation states in the ranges of oxidation reduction potentials (Eh from 0 6 V to −0 2 V) and pH [4−9] found in most environments [56] The actinide elements display similar behavior when they are in the same oxidation state, but, chemical characteristics can be markedly different if oxidation states are changed [26] Those changes in chemistry are very important for plutonium, neptunium and uranium because they display multiple oxidation states in aqueous solutions within the natural range of conditions The oxidation states for those elements in the environment may be U(IV), U(VI), Np(IV), Np(V), Pu(III), Pu(IV), Pu(V) and Pu(VI) The stability of their oxidation states varies considerably as demonstrated by the Eh-pH diagrams for uranium (Fig 2) [57] and plutonium (Fig 3) [58] Uranium is capable of maintaining the more soluble uranyl (+6) species at lower Eh values within the natural pH range than is plutonium in maintaining the plutonyl species (+6) However, over a fairly large Eh-pH region PuO_2^+ (+5) appears to be stable The higher oxidation state for neptunium, NpO_2^+, is intermediate in stability to those of uranium and plutonium and reduction to Np(IV) begins at an Eh of about 0 3 V at pH 6 [59] which means that Np(V) can exist over much of the normal environmental range of Eh and pH

Stability field diagrams are useful for establishing the boundary parameters for the existence of chemical species in the environment but they are limited in predictive capability because they can only be valid for the conditions under which they were measured The presence of complexing ligands and competing reactions in environmental media may modify the predicted oxidation state of actinides in soils and natural waters However, americium and curium appear to remain in the (+3) oxidation state over the normal range of environmental conditions

Measurements in the Irish Sea [60], Lake Michigan [61], North Pacific [62] and a Marshall Islands lagoon [63] show that 50 to 90 percent of the dissolved plutonium in oligotrophic water (low in plant nutrients, high in oxygen), with pH greater than 7, exists as an oxidized form The lower percentage is typical of measurements made at the surface and at intermediate depths in the open ocean The higher percentages were measured in Lake Michigan, the Irish Sea near Windscale, near the deep ocean bottom, and in lagoon water where interactions with sediment are more likely. NELSON and ORLANDINI [64] adapted a method developed by INOUE and TOCHIYAMA [65] for determining the presence of Np(V) to measure Pu(V) at environmental concentrations, they demonstrated that the oxidized form in several natural waters was in fact PuO_2^+ BONDIETTI, using a different procedure, produced confirmatory evidence for the existence of Pu(V) in a pond at Oak Ridge [66] Calculations based on the data in Table 4 suggest that Pu(V) is present in aqueous environments at concentrations of 10^{-13} to 10^{-17} M

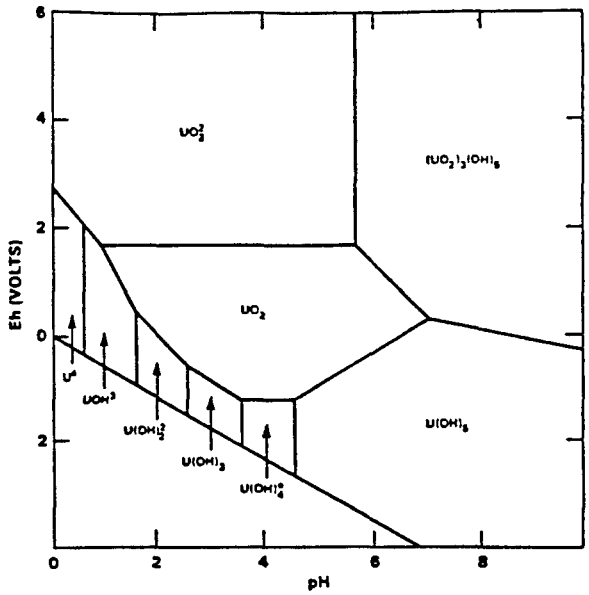

Fig 2 Eh pH diagram for uranium oxidation states at an equilibrium temperature of 25°C and total uranium concentration of 10^{-6} M (ref [57])

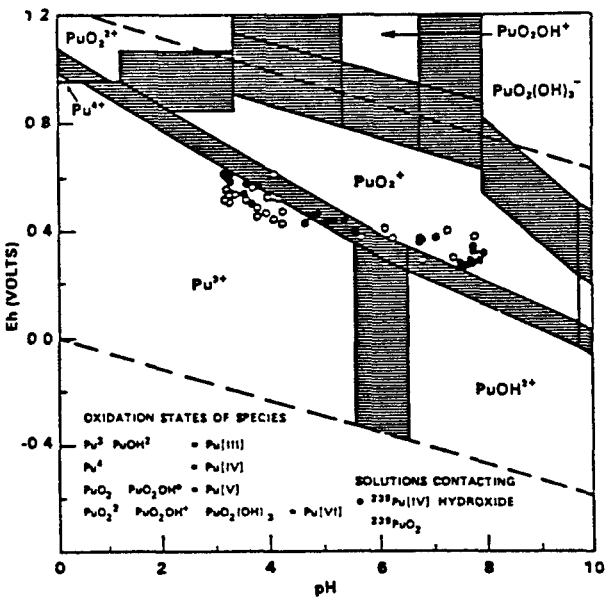

Fig 3 Equilibrium diagram of Eh pH at 25°C showing stability fields of different aqueous Pu species Dashed lines represent one atmosphere O_2 (upper) and one atmosphere H_2 (lower) Shaded areas represent regions of known uncertainty Data points represent Eh values measured at given pH for Pu(IV) hydroxide and PuO_2 (crystalline) in control with 0 0015 M $CaCl_2$ (ref [58])

Actinide concentrations often will be increased in water due to the presence of ligands that produce soluble complexes Those complexes may compete with redox and hydrolysis reactions that result in sorption of the actinide to soil and/or sediment It is clear that carbonate complexes of UO_2^{+2} enhance uranium solubility The formation of soluble complexes likely accounts for higher plutonium concentrations in some lake water compared with water from oligotrophic lakes (Table 6) [67] Higher concentrations in natural waters may be due to ligands such as sulfate ion or functional groups associated with dissolved

5

Table 6 *Comparison of plutonium concentrations and chemical variables in freshwater lakes* [67, 68, 69]

Lake	pH	DOC	$SO_4^=$ μg/l	Pu $ML^{-1} \times 10^{17}$
Lake Michigan	8 2	Oligotrophic	16	2 1
Last Mountain (Saskatchewan)	8 4	Oligotrophic	1300	4 5
Little Manito (Saskatchewan)	8 0		10^5	47
Banks (Georgia)	3 9			20
Okeefenokee (Florida)	5 0	35 ppm		15 4
Mono Lake	>9 0	high		130

organic carbon (DOC) The enhanced solubility of Pu, which has been observed in certain waters, seems to be best correlated with DOC NELSON *et al.* [68], in a summary of available field data, showed an inverse relationship between the sediment to water distribution ratio, K_d, and the concentration of DOC Further experiments, in which the amount of natural DOC was varied in the water, confirmed the inverse dependency of K_d on DOC and demonstrated that the binding capacity for plutonium may vary between sources of DOC Measurements at Mono Lake (California), which has a pH > 9 and is high in carbonate ion, has an elevated plutonium concentration in water, however, the oxidation state of this material has not yet been determined [69] The conditions in the lake appear to be appropriate for the presence of PuO_2^{+2} as a soluble carbonate complex

The sorption of actinides on sediments and soils represents one of the most important processes related to environmental mobility The stability with which actinides are bound to sediment and soil particles determines their concentration in natural waters, the relative importance of leaching and erosional processes in their movement in watersheds, and the ease with which they can transfer to plants through root uptake Sorption processes are complex in environmental media and may include various chemical reactions such as ion exchange reactions on soils and organic matter, surface adsorption mechanisms, oxidation reduction reactions and coprecipitation phenomena In a series of experiments using pond sediments contaminated with ^{238}Pu, EDGINGTON *et al* [70] demonstrated that the sorption-desorption of ^{238}Pu in contact with Lake Michigan water was an equilibrium process

Sorption of actinides with soils and sediments is significantly affected by the oxidation state as shown by studies with clay (Table 7) [26] and ocean sediments (Table 8) In the absence of stabilizing ligands, actinides in the lower oxidation states are more readily removed from aqueous solutions than those in the higher oxidation states because of their high ionic charge and tendency to hydrolyze to sparingly soluble forms

The mobile forms in soils and sediments, where both organic matter and microbial activity produce a low oxidation reduction potential, are likely to be the (III) and

Table 7 *Sorption of actinides to Miami silt loam clay* [26]

Actinide	% Sorbed[a]	K_d[b]
$^{233}Pu(IV)$	99 9	300000
$^{233}U(VI)$	95 6	4400
$^{237}Np(V)$	61 8	320

a At pH 6 5 and in 5 mM $Ca(NO_3)_2$ solutions

b $K_d = \frac{\text{conc on sorbent}}{\text{conc in solution}}$, units $ml^{-1}g$

Table 8 *Distribution coefficients (K_d) for* Pu (III *and* IV) *and* Pu (V *and* VI) *in windscale suspended sediments* [60]

Sample station	Pu (III + IV)	Pu (V + VI)
1	$3\ 4 \times 10^5$	8×10^3
2	2×10^6	$1\ 4 \times 10^4$
3	1×10^6	9×10^3
4	6×10^6	6×10^3
5 surface	$1\ 7 \times 10^6$	$1\ 4 \times 10^4$
5 bottom	3×10^6	$2\ 8 \times 10^4$
6	$5\ 4 \times 10^6$	–
7	5×10^5	$2\ 5 \times 10^4$

(IV) states NELSON and LOVETT [60, 71] determined that the plutonium adsorbed on sediments in the Irish Sea near Windscale was in the reduced state and that most of the Pu in interstitial water was also in the reduced state DAHLMAN *et al* [26] in experiments with fulvic acid extracted from soil demonstrated the reduction of Pu(VI) and the stabilization of soluble species of Pu(IV) WILDUNG and GARLAND [72] provided evidence that soil microflora produce soluble complexes with reduced plutonium through direct metabolic processes or indirectly by combination with microbial metabolites which may be present in soil

Physical transport processes

The importance of wind and water as transport mechanisms for actinides results from the fact that soil is a major repository of these elements in terrestrial ecosystems and that these elements are strongly sorbed to soils [73] Consequently, processes which transport soil have a direct impact on transport of soil associated contaminants Past studies have suggested a strong relationship between contaminant concentration and soil particle size for agricultural chemicals [74 75] and for plutonium [76, 77, 78] Because specific surface area (m^2 per g of soil) increases markedly with decreasing particle size [79] and because different chemical associations are influenced by particle size [80], much higher contaminant concentrations are usually associated with particles in the silt-clay size range than with larger particles Wind and water driven erosional processes result in particle sorting [81, 82] The combination of particle sorting by erosional processes and the differential association of contaminants by sediment particle size produce complex relationships for contaminant transport in terrestrial ecosystems The physics of sand trans-

port by wind is described in detail by BAGNOLD [81] and climatic factors affecting wind erosional processes, especially in arid and semiarid areas, are described by MARSHALL [83] Prediction of actinide transport by physical processes is not a well developed technology at the present time because of the highly variable nature of the driving force and a lack of understanding of the relationships that govern these processes Studies of plutonium transport by wind in arid and semi arid regions are reported elsewhere [84, 85, 86] Recent advances in the ability to predict plutonium transport by hydrologic processes will be discussed further to illustrate relationships that govern physical transport of actinides

Hydrologic transport processes of particular importance in the physical transport of soil-associated actinides include soil detachment by raindrop splash [87, 88, 89] and soil detachment and transport by overland flow [90, 91, 92] Soil particles detached by raindrop impact are important because they can be deposited on vegetation surfaces and thus provide a pathway for movement of soil associated actinides to plants (see section on transport to vegetation) Sediment transported by overland flow is important because it can redistribute contaminants and also deliver them to stream channels for subsequent transport to downstream areas

The combined phases of runoff, erosion, sediment transport, and deposition on upland areas and in stream channels usually result in enrichment of smaller sediment particles and organic matter in the transported sediment [82] and enrichment in concentration of sediment associated contaminants [74, 75, 93] This enrichment, which results from particle sorting and differential association of contaminants by particle size, is often expressed as an enrichment ratio the concentration of contaminant in the transported sediment divided by its concentration in the residual or uneroded soil Enrichment ratios tend to increase as the amount or rate of soil erosion (or runoff velocity) decreases [74] and have been related by regression analysis to sediment concentration, sediment discharge rate, and sediment yield [74, 75] By analyzing sediment transport rates by particle size classes in alluvial channels, LANE and HAKONSON [93] derived the following analytic expression for enrichment ratio in alluvial channels

$$ER = \frac{\Sigma\, Cs(d_i)\, Qs(d_i)}{Cs\, \Sigma\, Qs(d_i)} \qquad (1)$$

where

ER = alluvial channel enrichment ratio,
Cs(di) = Concentration of contaminant in sediment particles of size class i, with representative diameter di in millimeters
Qs(di) = Sediment transport (mass/time) for particles in size class i, with representative diameter di in millimeters
Cs = Mean concentration of contaminant over all particle size classes

Thus enrichment ratio was shown to be a variable and to be a function of contaminant concentrations in the soil by particle size classes, the particle size distribution, and the sediment discharge rates by particle size classes Equation 1 supports the empirical observation that enrichment ratio increases with decreasing sediment discharge rates For example, at very low sediment discharge rates (those associated with low runoff velocities) the bedload (coarse sediment particles) discharge rate is low and most of the transported sediment is in the smaller particle size classes Under those conditions, ER in Equation 1 would approach the ratio of concentrations in the finest size classes (Cs(di)) to the mean concentration over all size classes (Cs) At high sediment discharge rates (those associated with high runoff velocities) more of the bed sediments are in transport In the limit, if all of the bed sediments were in transport in the same proportion as they exist in the bed material, ER in Equation 1 would be unity

Field measurements of enrichment ratios for several elements at several locations in the United States are listed in Table 9 The first four entries in Table 9 represent enrichment ratios for soil nutrients in runoff from small agricultural areas, mean values vary from 2 6 to 7 1 The next two entries in Table 9 represent enrichment of fallout plutonium in runoff from small agricultural watersheds, values range from about 1 to 4 The last two entries in Table 9 represent enrichment of plutonium in runoff in stream channels representing larger watersheds Enrichment ratios observed at Los Alamos ranged from 1 4 to 13 3 with a mean of 5 5 Based on Equation 1, predicted enrichment ratios for Los Alamos stream channels ranged from 2 9 to 7 0 with a mean of 5 2 The rather close agreement between observed and predicted enrichment ratios suggests that particle sorting alone can account for the enrichment ratios observed at Los Alamos Although other factors undoubtly influence the observed enrichment ratios, recent analyses [75, 79, 93] suggest that particle sorting alone can produce enrichment ratios on the same order as those that are observed under field conditions In spite of wide differences in watershed size, hydrologic regime, and chemical characteristics inherent in the data in Table 9, enrichment ratios resulting from sediment transport are quite similar for several sediment associated contaminants Although transport processes undoubtedly differ between locations and contaminants, we suggest that particle sorting is one of the important factors involved in transport of sediment associated contaminants

Transport of biota

The importance of actinides as environmental contaminants depends on whether these materials enter biological pathways, and if so, do these pathways lead to man Considerable insight has been obtained on the behavior of plutonium and to a lesser extent, americium in terrestrial ecosystems The postulated behavior of the other actinides in the terrestrial environment is weakly supported by a few laboratory studies and by essentially, no field studies

7

Table 9 *Approximate enrichment ratios for nutrients and plutonium associated with sediment at various locations in the United States*

Land use and location	Approximate enrichment ratios mean	range	Comments	References
Cropland, USA[a]	4.5	2.5 – 7 4	Nitrogen	75
	3 6	2 6 – 6 0	Phosphorus	75
Rangeland, USA[a]	2 6	1 1 – 6 7	Nitrogen	75
	7 1	2 7 – 17	Phosphorus	75
Cropland, USA[b]	1 6	1 1 – 2 5	Fallout Plutonium	94
Pasture, USA[b]	2 3	0 8 – 4 0	Fallout Plutonium	94
Mixed Cropland, USA[c]	2 5	1 2 – 4 0	Fallout Plutonium, Transport in Perennial River	95
Semiarid, USA[d]	5.5	1 4 – 13 3	Waste Effluent Plutonium, Transport in Ephemeral Streams	93, 96

[a] Small agricultural watersheds (5 2–18 ha) at Chickasha, Oklahoma
[b] Small agricultural watersheds (2 6–2 9 hg) near Lebanon, Ohio
[c] Great Miami River (Drainage area = 1401 km²) at Sidney, Ohio
[d] Los Alamos Watersheds (176–15,000 ha) near Los Alamos, New Mexico

By nature of the wide diversity of conditions under which plutonium studies have been conducted in terrestrial ecosystems, we are in a position to evaluate the behavior of this element under a wide spectrum of study site and plutonium source conditions Climatic conditions ranging from and to humid and involving plutonium from weapons fallout, industrial waste effluents and accidental spills have been investigated [73]

Transport to vegetation

The two processes controlling actinide content of terrestrial plants are

physiological availability to plant roots with subsequent translocation to plant parts, and

the deposition of particles on foliage surfaces with or without subsequent absorption into plant tissues

The physiological availability of plutonium, americium and to a lesser degree, curium, and neptunium have been studied under a variety of controlled laboratory conditions [97, 98, 99, 100]

Based upon theoretical considerations the postulated availability of the actinides to plants [6, 101] is as follows

$$Np(V) > U(VI) > Am(III) \approx Cm(III) > Pu(IV) \approx \approx Th(IV) \approx Np(IV)$$

Pot culture studies tend to support that order of plant availability of the actinides to plants [6, 101] is as follows tion in plant over that in soil [97] The concentration ratio as applied to the actinides has been defined [102] as

$$\frac{\text{Activity/mass of receptor}}{\text{Activity/mass of donor}}$$

In terrestrial systems the donor compartment is usually considered to be soil Concentration ratios range from 10^{-4} to 10^{-8} for plutonium and 10^{-1} to 10^{-7} for americium Limited data for other actinides show concentration ratios of 10^{-3} to 10^{-4} for curium and 10^{-1} to 10^{-2} for neptunium

The physiological availability of any one actinide can be increased or decreased by at least an order of magnitude by soil amendments and indigenous soil factors For example, the addition of a chelating agent (DPTA) generally increases availability of plutonium and americium to plants, while liming treatment of the soil has been shown to reduce plant availability of americium [97] The importance of understanding the influence of soil amendments and indigenous soil factors on actinide availability to plants results from the fact that wastes from nuclear industries and fertilizers applied to agricultural lands often contain chemicals (e g chelators) that can modify actinide mobility [97]

Reduced plutonium that passes the root membrane migrates to the shoots of plants in the xylem in association with organic ligands [72] Differences in the gastrointestinal absorption of plutonium deposited in leaves versus stems, suggest that the chemical form of plutonium in these two plant tissues differ [72] Based on pot culture studies, available data on actinide distributions in various plant parts suggest that concentration patterns are as follows

$$\text{roots} > \text{leaf stem} > \text{seed fruit}$$

Concentrations ratios (plant/soil) of plutonium, americium, curium, and neptunium in seed fruits generally average from 1 to 2 orders of magnitude lower than the ratio for stem leaves [4, 98, 99] Thus, in situations where root uptake predominates in actinide movement to agricultural crops, food chain transport of the actinides to humans through ingestion of fruits and seeds will be diminished

Despite the host of chemical, biological and physical factors which can modify the physiological availability of actinides and subsequent transport within plant tissues [72, 97, 103], field studies in contaminated sites suggest

8

Table 10 *Comparison of plutonium concentration ratios for field and glasshouse conditions* [51]

Soil source	Field	Glasshouse
NTS[a] Area 11B	1.3×10^{-3} to 1.6×10^{-1}	1.5×10^{-4}
NTS Area 11C	4.5×10^{-3} to 3.4×10^{-1}	1.8×10^{-4}
NTS Area 13	7.8×10^{-3} to 4.4×10^{-1}	1.1×10^{-4}

[a] NTS (Nevada Test Site)

that contamination of foliage surfaces with particles containing actinides is the dominant transport mechanism in the environment under many conditions of climate for varied sources Comparative studies of plant uptake of plutonium under both field and laboratory conditions generally yield the relationships shown in Table 10 Studies focused specifically on root uptake of plutonium from soils yield plant soil concentration ratios which are at least one order of magnitude lower than the ratios observed under comparable conditions at field sites Those differences in concentration ratios imply that a mechanism exists in the environment for delivering at least 10 times more plutonium to vegetation than transport across root membranes The higher ratios observed at field sites are generally attributed to the presence of surficial contamination on field site vegetation That conclusion is supported by the obvious presence of soil on foliage surfaces and by the ability to remove some of the plutonium contamination from vegetation by washing [34, 104]

Plutonium from a reprocessing plant atmospheric effluent serves as the major source of contamination on adjacent vegetation at a site in the humid southeast U S [105] However, in most terrestrial sites contaminated with plutonium, direct fallout sources of plutonium are minimal relative to terrestrial sources such as wind and water resuspended soil

Studies in semi arid regions of New Mexico demonstrated that rain-splash of soil particles with subsequent deposition on foliage surfaces can contribute essentially all of the plutonium measured in field site vegetation [47] More importantly, those studies, which employed a labeled soil particle technique and the scanning electron microscope, have shown that relationships that govern translational movement of plutonium by erosion processes in soil also govern transport of plutonium to foliage surfaces For example, impacting raindrops caused an enrichment of small soil particles (< 105 μm) on foliage surfaces In general, only the highly transportable silt-clay particles (< 53 μm diameter), which generally contain higher concentrations of plutonium [106], are retained by plant surfaces Calculations based on the mass and plutonium content of soil measured on plants demonstrated that the rainsplash mechanism could easily account for the plutonium concentration ratios of 5×10^{-2} that were observed at this field site [34]

The absorption of plutonium through leaf surfaces has been demonstrated [103] but is considered to be a low order process in contaminated field sites particularly with annual or deciduous vegetation species

Studies on the uptake of plutonium by vegetable crops grown in contaminated field sites show that as much as 50% of the plutonium in crop samples was surficial contamination that could be removed by standard food preparation procedures [34] Plutonium that cannot be removed from vegetable crop surfaces by washing does not necessarily reflect plutonium incorporated into plant tissues CATALDO and VAUGHN [103] have shown that submicron particles on foliage surfaces are extremely difficult to remove by either simulated wind or rain

Under large scale agricultural conditions, a major source of actinides in crops may result from harvesting practices For example, most of the plutonium in grain grown in a contaminated field site at Savannah River Laboratory was attributed to cross-contamination with dust generated during mechanical threshing [24]

Transport to animals

The transport of actinides to animals is governed by the same processes that control the transport of these elements to plants That is, the actinides can be incorporated into animal tissues and/or they can be deposited on tissues exposed directly to the environment (i e lung, pelt-skin, and gastro-intestinal tract)

Based upon theoretical considerations and the assumption of similar conditions in the gut, actinide absorption from the gut [67] should follow

$$Np(V) > Cm(III) \approx Am(III) > Pu(IV)$$

Laboratory experiments on the physiological availability of actinides to animals generally follow the pattern listed above, plutonium is least available to animals, americium and curium are intermediate in availability, and neptunium is most available [10] However, many factors influence gut availability of the actinides such that rankings of availability are not rigid [67] For example, Pu(IV), which is complexed by microbial and/or plant tissues, may be more available for gut absorption than uncomplexed plutonium [107, 108] while P(VI) added to the gut containing food residues is reduced to P(IV) and thus becomes less available for gut absorption [109] The current ICRP [8] recommendation for gut absorption of plutonium by man is 1×10^{-4}

The concentrations of plutonium in animals collected from field sites indicates that gut availability of this element in the environment is low as shown by the low concentrations in internal organs and tissues In addition, highest concentrations of plutonium are invariably measured in tissues exposed to contamination with soil particles Plutonium in the pelt, gastro intestinal tract and to a lower degree, lungs accounts for nearly all of the animal's body burden [35, 50, 110]

Various assessments of the critical pathways of plutonium movement into man suggest that inhalation is the dominant pathway contributing to internal tissue dose [26, 111] While ingestion contributes as much as 10 times

9

more plutonium to man than inhalation, the low gut absorption (based on ICRP [8]) reduces the significance of the ingestion pathway in contributing to internal dose However, the recommended gut absorption factor of 1×10^{-4} [8] is based on laboratory studies with rats that were fed plutonium In light of recent concerns [112], a closer examination of gut absorption values for plutonium and other actinides, under environmental conditions is needed

The high mobility of large herbivores coupled with natural elimination processes provides a mechanism for actinide transport across the landscape Studies in nuclear fallout areas at Nevada Test Site [113] and in a nuclear spill area at Rocky Flats, Colorado [104] show that large herbivores (deer and cattle) ingest substantial quantities of plutonium-contaminated soil (i e several hundred grams per day for range cattle) Although the amount of plutonium transported across the landscape by this mechanism is considered to be small in areas where the extent of contamination is large (i e fallout areas) relative to the home range of the animal, there are circumstances where this transport mechanism becomes important For example, in a nuclear waste burial site at Hanford Washington, jack rabbits (*Lepus californicus*) which gained access to buried waste, ingested radioactive salts and subsequently excreted the salts on the surface of the site and surrounding area [114]

Studies on pocket gophers (*Thomomys bottae*) inhabiting a low-level waste site at Los Alamos, New Mexico [115], show that the burrowing activities of this animal can greatly perturb cover profiles placed over low-level radioactive waste disposal trenches Over a one year period, gophers excavated about 11 metric tons of soil per hectare from within the trench cover and created about 3000 m of tunnel system in the cover profile Animal burrowing activities can alter actinide distributions within the soil profile, as has been shown for pocket gophers in contaminated sites at Rocky Flats, Colorado [116] and for other small mammals at the Radioactive Waste Management Complex at Idaho National Engineering Laboratory [117]

Burrowing activities by animals can have a significant effect on the structure and output of dose assessment models for the actinides For example, at low-level waste sites, that use shallow-land burial methods, radionuclides brought to the surface along with soil casts become subject to physical transport processes as well as to physiological processes associated with root uptake As discussed previously, physical processes transfer at least 10 times more plutonium to vegetation than do physiological processes

Studies with honeybees at Los Alamos [118] demonstrated that small amounts of plutonium present in treated liquid wastes used by bees appear in honey Considering that most of the plutonium in the effluent is associated with particles ($< 30\%$ is associated with the fraction $< 0.05\ \mu m$ [119]), honeybees may be capable of transporting actinides that are both in solution and in association with particles in a liquid source to the honey

Based upon the analysis of available concentration ratios for the actinides, both chemical and physical processes contribute to the contamination of biological components of ecosystems Physical processes that cause soil to be transported to plants and animals dominate in the transport of plutonium and perhaps to a lesser degree, americium through food webs Although the plutonium and americium passing through an animal may be largely associated with particles, the relative importance of this source of actinide compared to that incorporated into food stuffs in contributing to internal tissue burdens is unknown However, it is a fact, in the case of plutonium, that physical processes can deliver at least 10 times more plutonium to plants than root uptake That fact would suggest the need to determine the relative importance of plutonium deposited on plants versus that incorporated into plants as a source of contamination to plant consumers

The general lack of data on curium, neptunium, uranium and thorium in terrestrial ecosystem components precludes any conclusions on food web transport All available data from laboratory studies indicate that those elements are more mobile than plutonium

Recent field studies at Oak Ridge [120, 121] show that the availability of actinides to plants and animals in two terrestrial study sites was

$$U > Cm \geqslant Am > Th \approx Pu$$

Uranium was about 10 times more available to plants and animals than plutonium and thorium Further field studies are needed to place bounds on the degree of transport of those elements to biota

Aquatic

Distribution and transport

Assessing the transport of actinides to aquatic biota by the conventional concentration ratio, as defined previously [102], presents a dilemma in that it is not always clear which aquatic component serves as the donor of the actinides to receptor components Concentration ratios for aquatic systems are usually based on water as the donor compartment Thus, CR's reported for plutonium suggest that aquatic organisms highly concentrate this element

As an example of the problem, a comparison of concentration ratios based on water versus those based on sediment as the donor compartment is given in Table 2 for several biotic components of Lake Michigan [122]

Regardless of how the ratio is calculated, it is clear that plutonium is attenuated as it passes through successively higher trophic levels Based upon observations in both freshwater and marine environments, it appears that concentration ratios decrease about one order of magnitude at each succeeding trophic level There is strong evidence

10

Table 11 *Comparison of plutonium concentration ratios for biological components of Lake Michigan using water versus sediment as the donor compartments* (Adapted from ref [122]

Compartment	Donor compartment Water[a]	Sediment
Mixed plankton	6300	5×10^{-2}
Benthic invertebrates	1300	1×10^{-2}
Zooplankton	250	2×10^{-3}
Benthic fish	250	2×10^{-3}
Planktivorous fish	60	5×10^{-4}
Piscivorous fish	0 5	3×10^{-6}
Water	1	8×10^{-6}
Sediment	130000	1

[a] concentration ratio = $\frac{\text{pCi Pu/g receptor}}{\text{pCi Pu/g donor}}$, data in table based on wet weights

that physical processes (i e surface attachment and/or ingestion of suspended particles and/or sediments) play an important role in contaminating aquatic biota with plutonium [112] Organisms living in close association with bottom sediments generally have the highest plutonium concentration ratios (Table 11)

The small amount of data on americium in aquatic biota do not provide a sufficient basis for comparison with corresponding plutonium data Some studies suggest that americium is more available than plutonium in the aquatic environs [123, 124] while other studies show no such patterns [125, 126] Field data on the other actinides appear to be completely lacking

Summary and conclusions

The chemical characteristics of the actinide elements cause them to be sorbed to soils and sediments to a large extent The stability of the sorption largely controls how mobile these elements will be in food chain processes such as root uptake in plants, gill transfer in fish, or ingestion via food or water

Although plutonium and americium are tightly bound to soils and sediments in the environment, a very small fraction of these elements is soluble and enters biological tissues The limited time-span (< 40 years) over which we have observed actinide behavior in the environment seriously limits our ability to forecast their behavior over the centuries and millenia during which many of these elements will be present in the environment However, preliminary observations on naturally occurring analog elements indicate that actinide solubility will likely not change appreciably with time

Present data demonstrate that soils and sediments serve as the major repository of plutonium in freshwater and terrestrial ecosystems and that processes which redistribute soils and sediment can also cause major changes in the environmental distribution of this element Although data-bases for the other actinides are small, physical processes will also provide a potentially important transport mechanism for these elements

It is clear that there is a need to determine the relative importance of actinides associated with soil and sediment as a source of contamination to biota Available data on actinides in terrestrial and fresh water ecosystems point to the potential importance of soil and sediment movement through food webs

Because physical transport processes operate at the soil-air or sediment water interface, changes in the distribution of plutonium within the soil sediment profile will alter the importance of this transport pathway Present distributions of plutonium in soil profiles from sites contaminated up to 35 years ago [35, 50] indicate that with time plutonium is depleted from the soil surface either from losses with eroding soil or from transport into the soil profile Whether those changes in plutonium distribution will change the relative importance of, what at present, are low order chemical processes is unknown In many aquatic systems, plutonium migration into the sediments through sedimentation and /or chemical processes may isolate the plutonium from the biosphere In terrestrial systems, losses of plutonium into the soil profile may create conditions more favorable for root uptake

Any phenomena which retain actinides in contact with the biosphere for extended times such as has been observed in arctic ecosystems will increase risks due to exposure to these elements The interception properties of vegetation cover, action by organisms living in the soil, and processes which resuspend sediments in aquatic ecosystems all contribute to maintaining actinides within the biosphere

With few exceptions, all present sources of actinides in terrestrial and aquatic ecosystems have resulted in very low transfer of these elements into food webs regardless of the transport process Doses to humans resulting from ingestion of food stuffs contaminated with the actinides have been uniformly low — much below doses incurred by humans from natural sources

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